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Development of the Molybdenum-Catalyzed Deoxydehydration of Polyols

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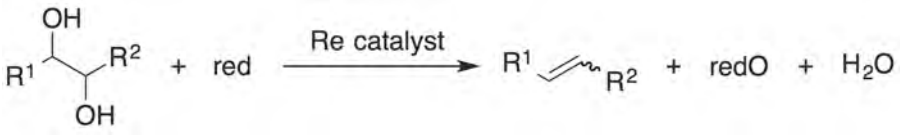
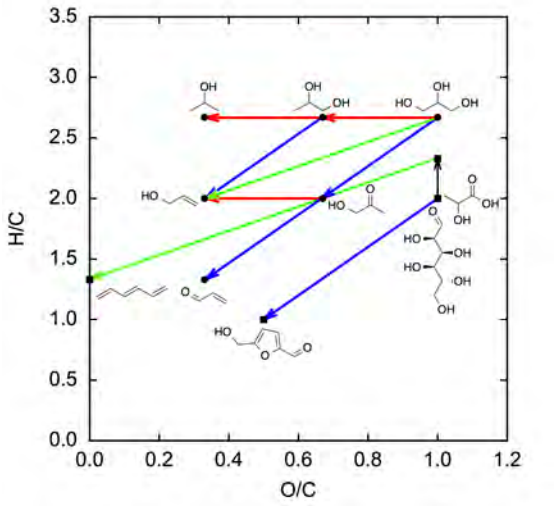
The transformation of biomass-derived compounds into platform chemicals is one of the possible contributions from chemistry to the realisation of an economy completely independent of fossil reserves. In order for oxygenrich biomass-derived compounds such as glycerol and sugar alcohols to substitute petroleum, reactions capable of reducing their oxygen content are in demand, and in particular the deoxydehydration (DODH) of vicinal diols into alkenes in the presence of a reductant ("red") and a catalyst (typically a rhenium complex) has received ample recent attention from the scientific community (figure 1).[1]



The transformation represents a model system for the various hydroxyl groups present in biomass (e.g., in carbohydrates), but is also relevant in itself as a potentially useful transformation of glycerol. The reaction is interesting since it combined a formal reduction and dehydration in a single chemical transformation. This makes the change in the van Krevelen diagram quite dramatic as exemplified by the triple deoxydehydration converting sorbitol to the completely deoxygenated 1,3,5-hexatriene (figure 2).



Considering the huge amounts of glycerol and sugar alcohols that would have to be processed for the utilization of biomass to make a substantial impact, the development of alternative catalysts, based on elements with higher terrestrial abundance than rhenium, is clearly desirable. We therefore set out to investigate the possibility of using the transition metal molybdenum because of the similarities between oxorhenium(VII) and oxomolybdenum(VI) complexes. In this talk our results from both experiments[2] and theoretical modelling using density functional theory will be presented.[3] Application of the methodology for the conversion of glycerol will be presented.

(1) Dethlefsen and Fristrup *ChemSusChem* **2015**, *8*, 767 (minireview).
(2) a) J .R. Dethlefsen and P. Fristrup *ChemCatChem* **2015**, *7*, 1184-1196. b) Dethlefsen, Lupp, Teshome, Nielsen, Fristrup *ACS Catalysis* **2015**, *5,* 3638-3647
(3) Lupp, Christensen, Dethlefsen, Fristrup *Chem. Eur. J.* **2015**, *21*, 3435-3432